



POCKET NO: 217721US0X PCT

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF

CARLO PEREGO, ET AL.

SERIAL NO: 10/019,273

FILED: APRIL 9, 2002

FOR: PROCESS FOR THE PRODUCTION
OF PROPYLENE FROM OLEFINIC
STREAMS

:

: EXAMINER: DANG, THUAN D.

:

: GROUP ART UNIT: 1764

APPEAL BRIEF

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

SIR:

This is an appeal of the Rejection dated May 6, 2005 of twice-rejected Claims 1-14 and 17-20. A Notice of Appeal is **submitted herewith**.

I. REAL PARTY IN INTEREST

The real party in interest in this appeal is Enitecnologie S.P.A. having an address Via F. Maritano 26 I-20097 San Donato, Milanese-Milan, Italy.

II. RELATED APPEALS AND INTERFERENCES

Appellants, Appellants' legal representative and the assignee are aware of no appeals or interferences which will directly affect or be directly affected by or have a bearing on the Board's decision in this appeal.

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III. STATUS OF THE CLAIMS

Claims 1-14 and 17-20, all the claims in the application, stand rejected and are herein appealed.

IV. STATUS OF THE AMENDMENTS

No amendment under 37 CFR 1.116 has been filed.

V. SUMMARY OF THE CLAIMED SUBJECT MATTER

Independent Claim 1 is drawn to a process for the production of propylene comprising contacting a mixture of hydrocarbons with a catalyst composition under cracking conditions, thereby producing a product comprising propylene from said mixture, wherein the mixture of hydrocarbons comprises predominately olefins, the mixture has a boiling point ranging from -15°C to +80°C, the catalyst composition comprises a large pore zeolite comprising a lattice of 12 tetrahedrons, and the zeolite has a molar ratio of silica/alumina from 100 to 200.

See the specification at page 5, lines 12-20 and page 9, line 4ff.

Independent Claim 20 is drawn to a process for the production of propylene comprising contacting a mixture of hydrocarbons with a catalyst composition under cracking conditions, thereby producing a product comprising propylene from said mixture, wherein the mixture of hydrocarbons comprises predominately olefins, the mixture has a boiling point ranging from -15°C to +80°C, the catalyst composition comprises a large pore zeolite

comprising a lattice of 12 tetrahedrons, and the zeolite has a molar ratio of silica/alumina less than 200; and wherein the zeolite is prepared by the steps comprising:

contacting sodium aluminate with an aqueous solution of tetramethyammonium hydroxide to form a mixture,

contacting the mixture with colloidal silica to form a homogeneous gel,

crystallizing the gel under hydrothermal conditions to obtain a first solid,

washing the first solid with water to form a second solid,

calcining the second solid in air to form a calcined solid,

subjecting the calcined solid to an ion exchange using an aqueous solution of ammonium acetate to form a third solid, and

calcining the third solid in air.

See the specification at page 5, lines 12-20, and pages 9-10 (Example 1).

VI. GROUNDS OF REJECTION

Ground (A)

Claims 1-14 and 17-20 stand rejected under 35 U.S.C. § 103(a) as unpatentable over U.S. 5,026,936 (Leyshon et al) in view of U.S. 3,832,449 (Rosinski et al).

Ground (B)

Claims 17-20 stand rejected under 35 U.S.C. § 112, first paragraph, as failing to satisfy the written description requirement thereof.

Ground (C)

Claims 17-19 stand rejected under 35 U.S.C. § 112, second paragraph, as indefinite.

VII. ARGUMENT

Ground (A)

Claims 1-14 and 17-20 stand rejected under 35 U.S.C. § 103(a) as unpatentable over Leyshon et al in view of Rosinski et al. That rejection is untenable and should not be sustained.

The present invention relates to a process for the production of propylene under cracking conditions by contacting a predominately olefinic hydrocarbon stream in the presence of a catalyst composition comprising a particular type of zeolite.

As described in the specification beginning at page 1, line 21, catalytic cracking processes for producing propylene are known, including such processes using zeolite catalysts. Even with these prior art zeolite catalysts, however, experts in the field still require greater conversions and at the same time, increased stability of catalytic activity over a period of time.

The presently-claimed process uses materials capable of improving the yield to propylene and that also have a great advantage of maintaining the catalytic performance practically constant over a period of time. Thus, as recited in Claim 1, the invention is a process for the production of propylene comprising contacting a mixture of hydrocarbons with a catalyst composition under cracking conditions, thereby producing a product comprising propylene from said mixture, wherein the mixture of hydrocarbons comprises predominately olefins, the mixture has a boiling point ranging from -15°C to +80°C, the

catalyst composition comprises a large pore zeolite comprising a lattice of 12 tetrahedrons, and the zeolite has a molar ratio of silica/alumina from 100 to 200.

ZSM-12 zeolite is a species of a large-pore zeolite, i.e., a zeolite having a lattice consisting of 12 tetrahedrons, as described in the specification in the paragraph bridging pages 6 and 7. As Applicants describe in the specification beginning at page 7, line 15, contrary to what is specified in scientific and patent literature, the ZSM-12 material has the best catalytic performance at $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratios < 200 (mol/mol).

The specification contains comparative data demonstrating both the significance of the molar ratio of silica/alumina, when using a ZSM-12 catalyst, and a ZSM-12 catalyst compared to a ZSM-5 catalyst, as now discussed. Example 3 demonstrates catalytic testing of a ZSM-12 zeolite having a molar ratio $\text{SiO}_2/\text{Al}_2\text{O}_3$ of 100. Comparative Example 4 is otherwise similar but employs a molar ratio $\text{SiO}_2/\text{Al}_2\text{O}_3$ of 250. The results for total conversion and selectivity to propylene are shown for Example 3 and Comparative Example 4 in Figures 1 and 2, respectively. The differences in results are manifest. For Example 3, and as described in the specification at page 12, lines 15-19, the total conversion and selectivity to propylene are high and remain steady for a time on stream (tos) of a least 140 hours, thus demonstrating unexpected stability and no catalytic deterioration phenomena during this time. For Comparative Example 4, on the other hand, and as described in the specification at page 15, lines 5-12, contrary to what is specified in the literature, the catalytic performance of ZSM-12 with a molar ratio $\text{SiO}_2/\text{Al}_2\text{O}_3$ of 250 is lower both in terms of yield and duration, with respect to the zeolite having a greater content of Al_2O_3 , and already after 25 hours of tos, evident catalytic deterioration phenomena are present.

Comparative Example 5 exemplifies a commercial ZSM-5 zeolite having a molar ratio $\text{SiO}_2/\text{Al}_2\text{O}_3$ of 30. As shown in Figure 3, and as described in the specification at page 17, lines 13-15, the catalytic performances of ZSM-5 are much lower both in terms of yield (product of selectivity and conversion) and duration, with respect to the ZSM-12 zeolite. Indeed, as indicated by Figure 3, and as described in the specification at page 17, lines 16-18, already after 10 hours of tos, catalytic deterioration phenomena are present.

The above-discussed results could not have been predicted by the applied prior art.

Leyshon et al discloses a process for preparing propylene by cracking a mixture of C_{4+} hydrocarbons in the presence of apparently **any** zeolite catalyst, under conditions that favor production of a product mixture containing ethylene and propylene, separating and recovering propylene from this product mixture, and then subjecting the ethylene to a metathesis reaction to form further quantities of propylene (column 1, lines 8-17). Leyshon et al lists a relatively large group of zeolite catalysts, including ZSM-11, ZSM-21, ZSM-38, ZSM-23, ZSM-35, phosphorus-containing zeolites, zeolite A, zeolite X, zeolite Y, zeolite ZK-5, zeolite ZK-4, and others (column 3, line 65 through column 4, line 30). While Leyshon et al also lists ZSM-12, citing Rosinski et al (column 4, lines 13-14), Leyshon et al prefer acid zeolites, and most particularly a ZSM-5 zeolite (column 4, lines 17-19). Indeed, a ZSM-5 catalyst is the only catalyst exemplified in an example of Leyshon et al.

Rosinski et al, as discussed above, discloses a particular ZSM-12 zeolite, for use in cracking and other catalytic processes, although cracking of a predominately olefin-containing hydrocarbon mixture to form propylene is not specifically disclosed therein. In addition, while Rosinski et al discloses a broad, preferred, and particularly preferred range of $\text{YO}_2/\text{W}_2\text{O}_3$, wherein W is Al or Ga, and Y is Si or Ge, Rosinski et al does not relate these

ratios to the effectiveness of the produced zeolite as a catalyst for a particular application. In view of the multitude of applications disclosed, these ratios are essentially meaningless.

It is only with the present disclosure as a guide that one skilled in the art would choose the ZSM-12 catalyst of Rosinski et al, and one having a molar ratio $\text{SiO}_2/\text{Al}_2\text{O}_3$ within the presently-recited range, in the process of Leyshon et al, given the fact that Leyshon et al is not limited with regard to the particular zeolite catalyst used, and considering the fact that Leyshon et al actually prefers a ZSM-5 catalyst, shown above as being inferior to a ZSM-12 catalyst. Indeed, Leyshon et al is so broad, as not even being sufficient to present a *prima facie* case of obviousness.

The fact that a claimed species or subgenus is encompassed by a prior art genus is not sufficient by itself to establish a *prima facie* case of obviousness. *In re Baird*, 16 F.3d 380, 382, 29 USPQ2d 1550, 1552 (Fed. Cir. 1994) (“The fact that a claimed compound may be encompassed by a disclosed generic formula does not by itself render that compound obvious.”) See also MPEP 2144.08.

In addition, since Leyshon et al requires a subsequent metathesis reaction, it suggests to persons skilled in the art that satisfactory yields of propylene require the addition of such a metathesis reaction. This is confirmed by the actual example (Example 1) in Leyshon et al. Example 1 shows a stream 103 following cracking and prior to subjecting the components of the stream to further processing, and then metathesis. As shown in Table 1 therein, stream 103 contains a total amount of product of 144.6 M pounds per hour, 30.4 pounds of which are propylene (wherein M is 1,000 pounds). The selectivity to propylene is therefore $(30.4/144.6) \times 100$, or 21%. This selectivity can be contrasted with the selectivity to propylene in above-discussed Example 3 of the present invention of about 40%, as shown in

Table 2 and Figure 1 herein. Note that the feed in Leyshon et al's Example 1, represented by stream 102, is not significantly different from the feed in above-discussed Example 3 of the present invention, shown in Table 1 herein at page 8, line 8ff of the specification.

Thus, in contradistinction to Leyshon et al's disclosure that the zeolites are generally interchangeable but that ZSM-5 is preferred, Applicants have shown that not all zeolites are interchangeable and that ZSM-5 is inferior to ZSM-12, and that the molar ratio of silica/alumina of the ZSM-12 used is significant.

Claims 3-4

To the extent the comparative data of record are not considered applicable to a large pore zeolite comprising a lattice of 12 tetrahedrons generally, which Applicants submit the data are, Claims 3 and 4 are separately patentable because the data have been shown specifically with ZSM-12 zeolite.

Claim 13

Claim 13 is separately patentable because, to the extent a person of ordinary skill in the art would combine Leyshon et al and Rosinski et al, even though Applicants submit that one skilled in the art would not, nevertheless, Rosinski et al directs persons skilled in the art away from the molar ratio of this claim, and to a particularly preferred ratio of 90-100 (Table 2 therein).

Claim 14

Claim 14 is separately patentable because, to the extent the above-discussed comparative data are not extrapolatable to other molar ratios, the actual data has been shown with the particular molar ratio of this claim.

Claims 17-19

Claims 17-19 are separately patentable, since the applied prior art neither discloses nor suggests maintenance of catalytic activity for the prescribed time recited in these claims.

Claim 20

Claim 20 is separately patentable, since the applied prior art neither discloses nor suggests preparing a zeolite having a molar ratio of silica/alumina less than 200 by the particular steps recited therein.

For all the above reasons, it is respectfully requested that the rejection over Leyshon et al in view of Rosinski et al be REVERSED.

Ground (B)

Claims 17-20 stand rejected under 35 U.S.C. § 112, first paragraph, as failing to satisfy the written description requirement thereof. That rejection is untenable and should not be sustained.

Claims 17-19

Applicants describe in the specification at page 12, lines 15-19, with regard to Figure 1 that no catalytic deterioration phenomena are present until at least 140 hours of tos, and at page 15, lines 10-12, with regard to Figure 2 that already after 25 hours of tos, evident catalytic deterioration phenomena are present. These disclosures together support the limitation “for 25 hours or more.”

Claim 20

While the relied on disclosure is for preparation of ZSM-12, ZSM-12 is a species of a large-pore zeolite, i.e., a zeolite having a lattice consisting of 12 tetrahedrons, which would be prepared in a similar manner, as would be well-understood by persons skilled in the art. Such a person would appreciate that Applicants were in possession of the invention of Claim 20 as of the appropriate filing date.

For all the above reasons, it is respectfully requested that this rejection be REVERSED.

Ground (C)

Claims 17-19 stand rejected under 35 U.S.C. § 112, second paragraph, as indefinite. That rejection is untenable and should not be sustained.

With regard to the term “25 hours or more”, the term “or more” is intended to cover an unlimited period, or at least as long as the composition maintains catalytic activity.

For all the above reasons, it is respectfully requested that this rejection be
REVERSED.

VIII. CONCLUSION

For the above reasons, it is respectfully requested that all the rejections still pending
be REVERSED.

Respectfully submitted,

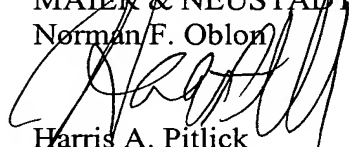
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EXHIBITS APPENDIX

Claim 1: A process for the production of propylene comprising contacting a mixture of hydrocarbons with a catalyst composition under cracking conditions, thereby producing a product comprising propylene from said mixture,

wherein the mixture of hydrocarbons comprises predominately olefins, the mixture has a boiling point ranging from -15°C to +80°C, the catalyst composition comprises a large pore zeolite comprising a lattice of 12 tetrahedrons, and the zeolite has a molar ratio of silica/alumina from 100 to 200.

Claim 2: The process according to claim 1, wherein the mixture of hydrocarbons has a boiling point ranging from -12°C to +60°C.

Claim 3: The process according to claim 1, wherein the zeolite is a ZSM-12 zeolite.

Claim 4: The process according to claim 3, wherein the ZSM-12 zeolite has a molar ratio silica/alumina ranging from 100 to 150.

Claim 5: The process according to claim 1, wherein the mixture of hydrocarbons comprises from 30% to 100% by weight of olefins.

Claim 6: The process according to claim 5, wherein the mixture of hydrocarbons has a content of 40% to 85% by weight of olefins.

Claim 7: The process according to claim 1, wherein the process is carried out at a temperature ranging from 400°C to 750°C.

Claim 8: The process according to claim 7, wherein the temperature ranges from 450°C to 700°C.

Claim 9: The process according to claim 8, wherein the temperature ranges from 500°C to 650°C.

Claim 10: The process according to claim 1, wherein the process is carried out at a weight hourly space velocity (WHSV) ranging from 0.1 h⁻¹ to 1,000 h⁻¹.

Claim 11: The process according to claim 10, wherein the weight hourly space velocity ranges from 0.5 h⁻¹ to 100 h⁻¹.

Claim 12: The process according to claim 11, wherein the weight hourly space velocity ranges from 0.8 h⁻¹ to 50 h⁻¹.

Claim 13: The process according to claim 1, wherein the zeolite has a molar ratio of silica/alumina ranging from 150 to 200.

Claim 14: The process according to claim 1, wherein the zeolite has a molar ratio of silica/alumina of 100.

Claim 17: The process according to claim 1, wherein the catalyst composition maintains catalytic activity for 25 hours or more.

Claim 18: The process according to claim 1, wherein the catalyst composition maintains approximately the same level of conversion for 25 hours or more.

Claim 19: The process according to claim 1, wherein the catalyst composition maintains catalytic activity and maintains approximately the same level of conversion, both for 25 hours or more.

Claim 20: A process for the production of propylene comprising contacting a mixture of hydrocarbons with a catalyst composition under cracking conditions, thereby producing a product comprising propylene from said mixture,

wherein the mixture of hydrocarbons comprises predominately olefins, the mixture has a boiling point ranging from -15°C to +80°C, the catalyst composition comprises a large pore zeolite comprising a lattice of 12 tetrahedrons, and the zeolite has a molar ratio of silica/alumina less than 200; and

wherein the zeolite is prepared by the steps comprising:

contacting sodium aluminate with an aqueous solution of tetramethyammonium hydroxide to form a mixture,

contacting the mixture with colloidal silica to form a homogeneous gel,

crystallizing the gel under hydrothermal conditions to obtain a first solid,

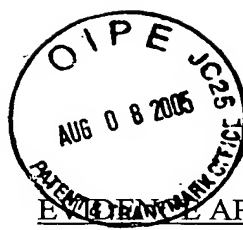
washing the first solid with water to form a second solid,

calcining the second solid in air to form a calcined solid,

subjecting the calcined solid to an ion exchange using an aqueous solution of ammonium acetate to form a third solid, and

calcining the third solid in air.

Application No. 10/019,293
Appeal brief



EVIDENCE APPENDIX

None.

Application No. 10/019,273
Appeal Brief

RELATED PROCEEDINGS APPENDIX

None.